

Electronic Supplementary Information

Vertical Recrystallization for Highly Efficient and Stable Formamidinium based Inverted Structure Perovskite Solar Cells

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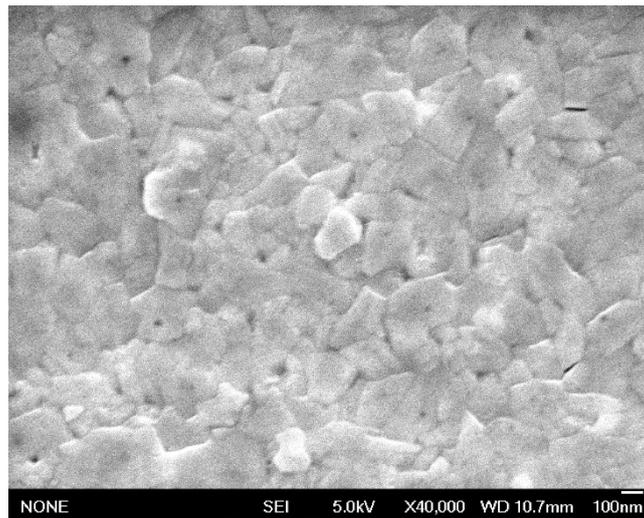


Figure S1. SEM image of FAPbI₃ film without annealing (δ -FAPbI₃ film). The crystal size is around 100 nm.

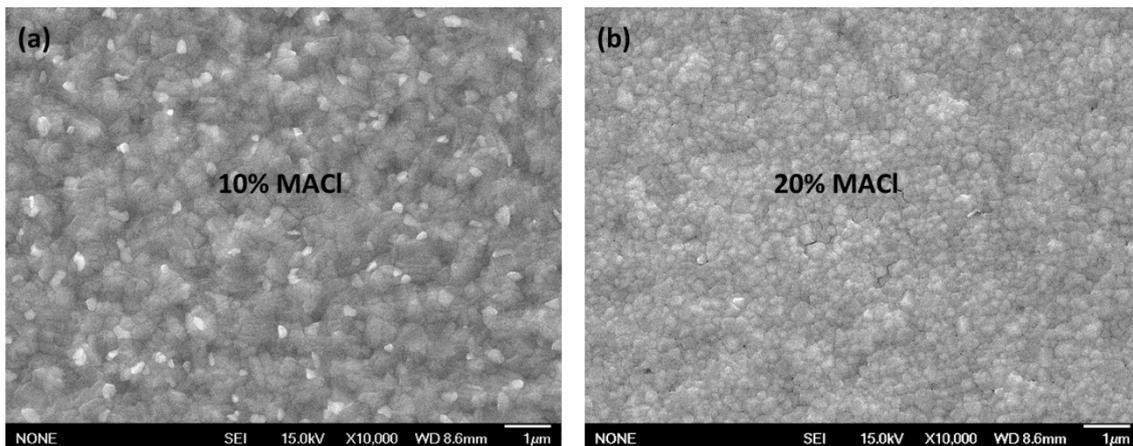


Figure S2. SEM image of perovskite film formed by mixed FAPbI₃+MACl in precursor solution. (a) FAPbI₃(MACl 10%), (b) FAPbI₃(MACl 20%). The perovskite films were thermal annealed at 100°C for 10 min and then annealed at 140°C for 20min.

Different from MACl vertical recrystallization method, the perovskite film formed by the mixed FAPbI₃+MACl method showed small perovskite grains (about 200nm), which is consistent with previous report.^[1]

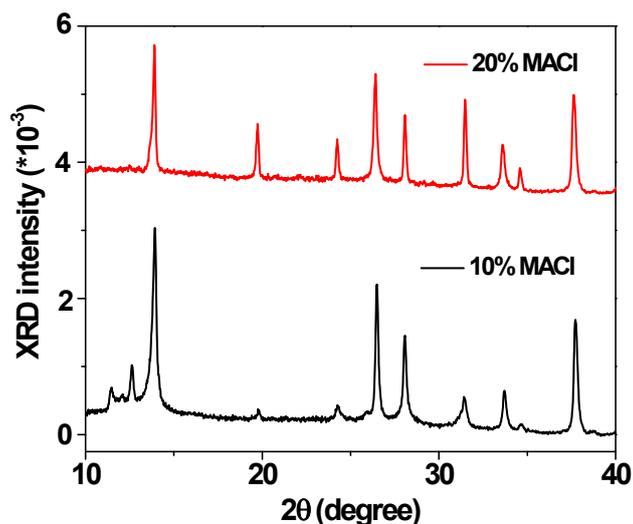


Figure S3. XRD spectrum of perovskite film formed by mixed FAPbI₃/MACl. (a) FAPbI₃(MACl 10%), (b) FAPbI₃(MACl 20%). The perovskite films were thermal annealed at 100°C for 10 min and then annealed at 140°C for 20min. Different from MACl vertical recrystallization method, the perovskite film formed by the mixed FAPbI₃+MACl method showed very weak XRD peak.

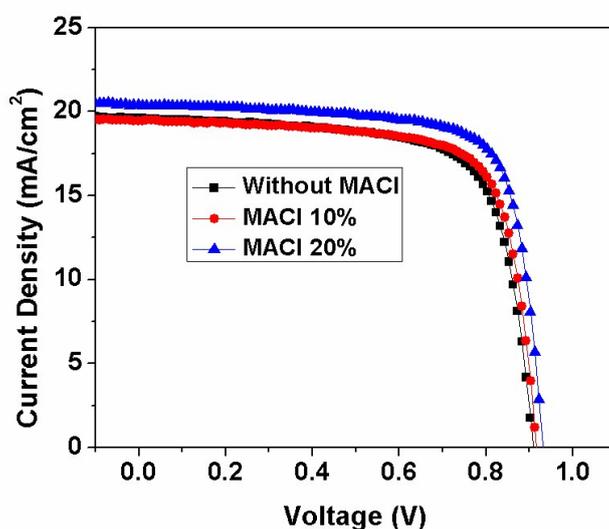


Figure S4. Current density-voltage (J-V) curves FAPbI₃ perovskite device using pristine-FAPbI₃ film (black square dots line), mixed FAPbI₃+MACl 10% (red rounded dots line), and mixed FAPbI₃+MACl 20% (blue triangle dots line). The perovskite films were thermal annealed at 100°C for 10 min and then annealed at 140°C for 20min.

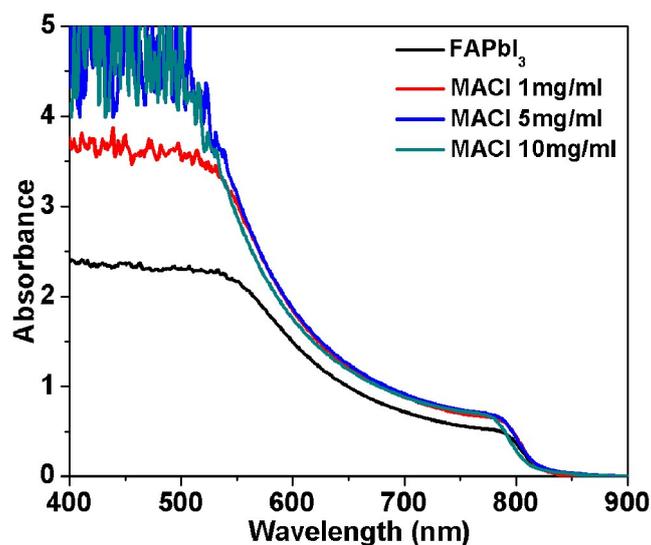


Figure S5. Absorbance spectra of FAPb₃ film with different MACl concentration treatment.

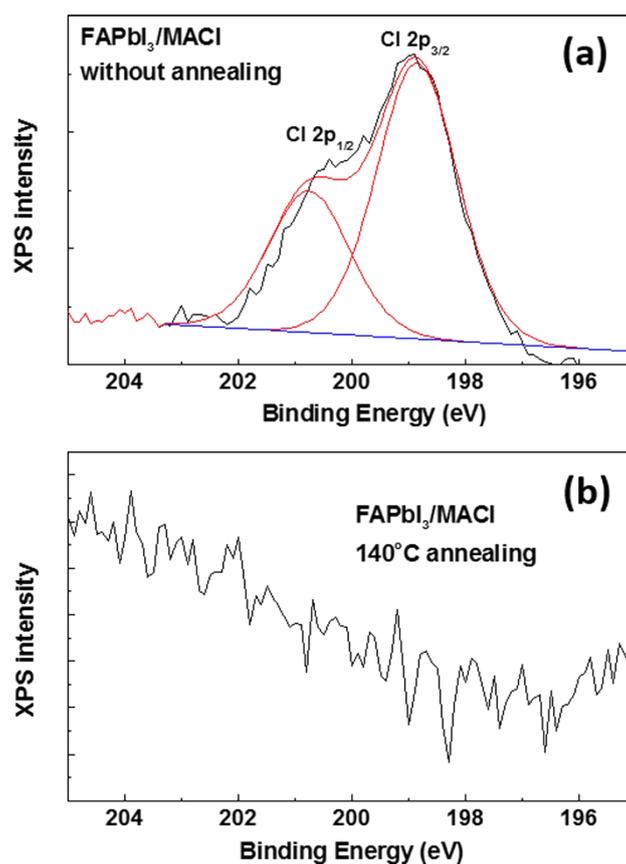


Figure S6. XPS spectra (Cl 2p core level) of (a) a FAPb₃ perovskite film with 10 mg/ml MACl spin-coated on top surface without thermal annealing. (b) a FAPb₃ perovskite film with 10 mg/ml MACl treatment after thermal annealing at 100°C for 10 min and 140°C for 20 min, indicated little amount of Cl was remained in the perovskite film.

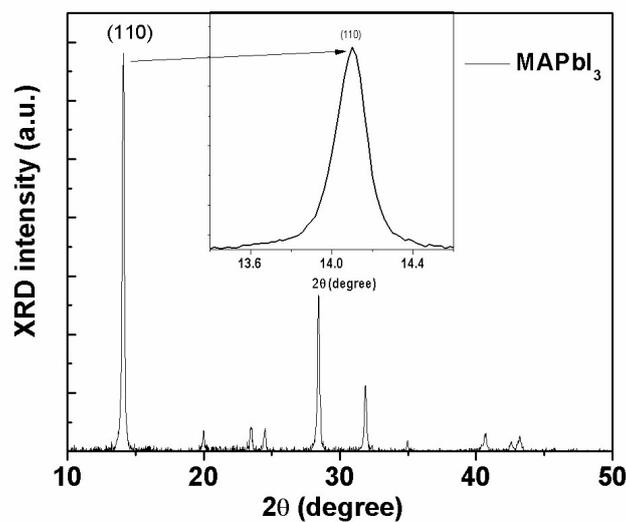


Figure S7. X-ray diffraction pattern of MAPbI₃ film.

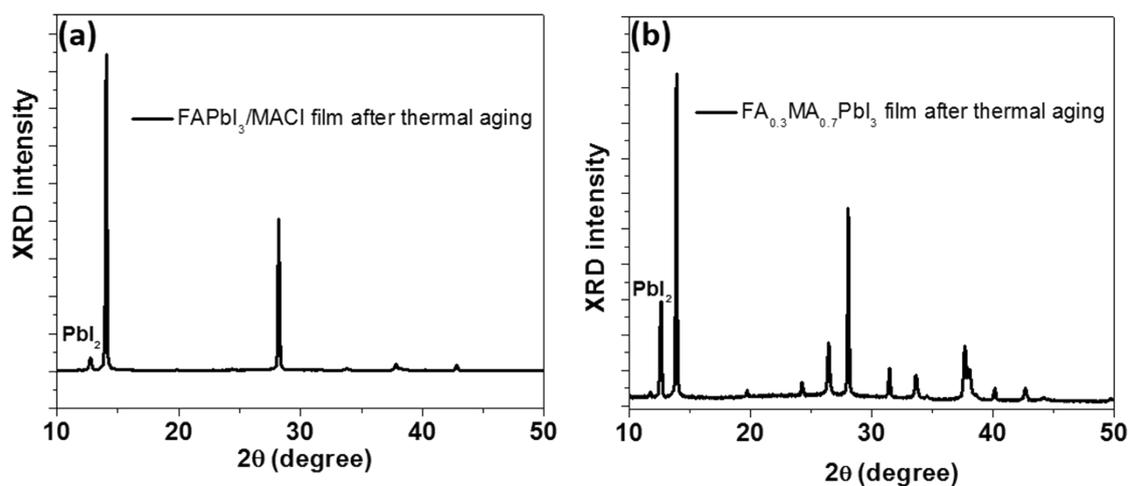


Figure S8. (a) XRD spectrum of thermal aged perovskite film (Vertical recrystallization method, FAPbI₃/MACI 5mg/ml). (b) XRD spectrum of thermal aged perovskite film (FA_{0.3}MA_{0.7}PbI₃ by reported method^[2]). Thermal aging of perovskite films were performed at 100°C on hot plate for 50 hours in glove box.

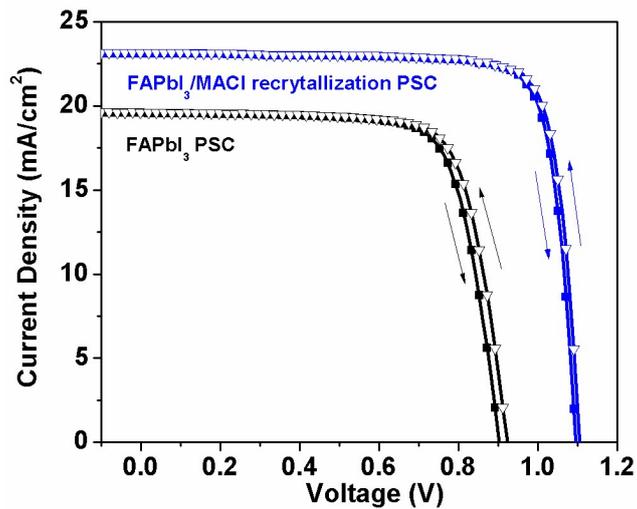


Figure S9 Hysteresis behavior of pristine FAPbI₃-based PSC and FAPbI₃/MACl (5 mg/ml) based inverted PSC.

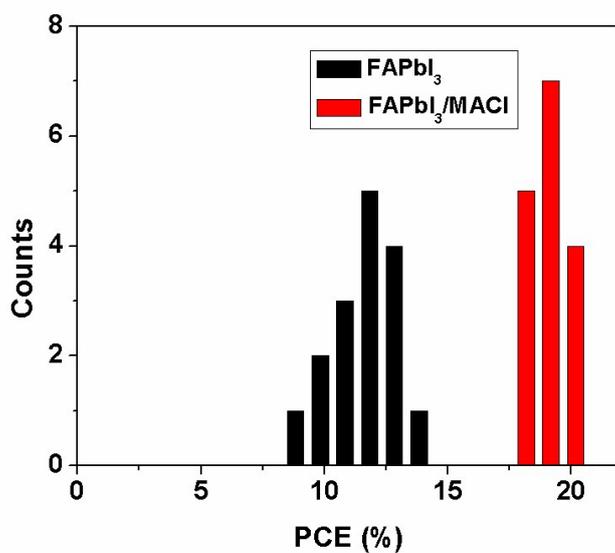


Figure S10. Statistical PCE of of the pristine FAPbI₃-based PSC and FAPbI₃/MACl (5 mg/ml) based inverted PSC.

Table S1. Photovoltaic data obtained from FAPbI₃ perovskite device with 0mg/ml, 1mg/ml, 5 mg/ml and 10mg/ml MACl treatment.

	V _{oc} (V)	J _{sc} (mA/cm ²)	FF	PCE %
without MACl	0.91	19.57	0.741	13.24
MACl 1mg/ml	1.02	20.49	0.751	15.72
MACl 5mg/ml	1.10	23.09	0.814	20.65
MACl 10mg/ml	1.09	22.51	0.811	19.89

Table S2. Summary of lattice parameters for perovskite films

Perovskite film	XRD (110)	lattice parameter
FAPbI ₃	13.86	6.384
1mg/ml MACl	13.88	6.375
5mg/ml MACl	13.90	6.366
10mg/ml MACl	13.96	6.339
MAPbI ₃	14.10	6.276

Reference:

- [1] Z. Wang, Y. Zhou, S. Pang, Z. Xiao, J. Zhang, W. Chai, H. Xu, Z. Liu, N. P. Padture, G. Cui, *Chemistry of Materials* 2015, 27, 7149.
- [2] J. Liu, Y. Shirai, X. Yang, Y. Yue, W. Chen, Y. Wu, A. Islam, L. Han, *Advanced Materials* 2015, 27, 4918.